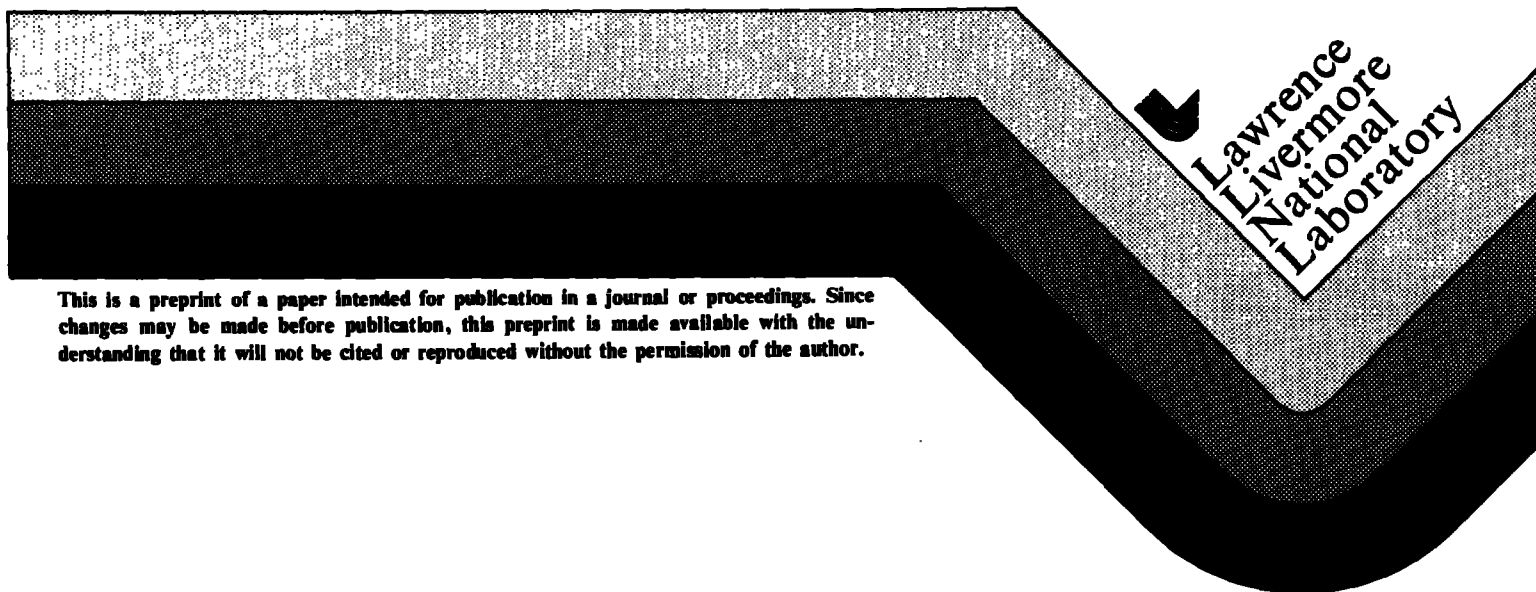# Krypton-85 Purge at Three Mile Island:
A Comparison of Measured and Calculated
Surface Air Concentrations

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This paper was prepared for submission to <u>Science</u>

September 1983

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Krypton-85 Purge at Three Mile Island: A Comparison of Measured
and Calculated Surface Air Concentrations*

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ABSTRACT

Krypton-85 was released from the Three Mile Island nuclear power station during a
planned purge of the containment system in the summer of 1980. Air concentrations
measured to distances of 63 km were compared to results calculated by the
MATHEW/ADPIC particle-in-cell model. Measurements and calculations agreed within
the error bounds of previous model evaluations.

Models are being developed to calculate the dispersion of radioactive gases accidentally discharged from nuclear power reactors and to estimate the resulting health risk. Protective action decisions will be based in part on these model predictions.

During the accident at the Three Mile Island (TMI) Unit No. 2 pressurized water reactor in March 1979, approximately 45,000 Ci of ^{85}Kr were trapped in the reactor building. Between 28 June and 11 July 1980 the ^{85}Kr was released through the plant venting stack at a controlled, measured rate. This purge offered an opportunity to test model estimates of the ^{85}Kr dispersion with actual measurements of surface air concentrations.

During the purge the Environmental Protection Agency (EPA) collected daily air samples for ^{85}Kr measurements at fixed locations within 8.5 km of TMI [1]. Lawrence Livermore National Laboratory (LLNL) used computer models in real time to generate hourly ^{85}Kr concentration-contour maps, which were transmitted to LLNL personnel near TMI.

To evaluate the LLNL models at greater distances, the Radiological Sciences Insitute (RSI) of the New York State Health Department measured ^{85}Kr concentrations on 8 July 1980, toward the end of the purge, at sites ranging from 36 to 63 km from the reactor (Fig. 1). Middlesex was upwind of TMI and was used as a control to assess background levels of ^{85}Kr. The measured concentrations were subsequently compared to the results of model calculations performed by LLNL [2].

On the morning of 8 July, venting at TMI was terminated shortly before 0630 EDT due to a faulty radiation monitor. At 1224 EDT venting was resumed, and for the next 12 nours the release rate of ^{85}Kr varied between 228 and 447 Ci/hour, with an average of 335 Ci/hour. Surface winds were from the western quadrant at speeds ranging from near calm to 10 meters per second. To allow the plume to reach the RSI sampling locations nearest the reactor, sampling was begun two hours after the resumption of venting.

At each of the seven RSI downwind locations, three samples were collected in evacuated, 35-liter metal containers fitted with needle valves and flow meters to regulate the inflow of air. Each container was used to sample air continuously for periods ranging from 1 hour 25 minutes to 3 hours 41 minutes. Most samples were collected in approximately 2 hours. The flow rate into the evacuated containers was initially set at 150 cm^3 per minute; it remained nearly constant during the first 1.5 hours of sampling and then dropped to approximately 100 cm^3 per minute at about 2 hours. For longer sampling times the flow rate decreased more rapidly. All samples were collected at a height of 1.5 meters above ground.

To measure the concentration of ^{85}Kr the RSI air sample was mixed with 2 cm^3 of Kr carrier. The Kr was then separated from the air by cryogenic adsorption on charcoal, followed by chromatographic purification. The concentration of ^{85}Kr was measured by internal gas-proportional counting (3).

MATHEW (4) and ADPIC (5), the combination of models used to simulate plume dispersion in this study, were developed as part of the Atmospheric Release Advisory Capability at LLNL. MATHEW is a three-dimensional, diagnostic, regional wind adjustment model that supplies ADPIC with mass-consistent velocity fields. Terrain (in block form) has been added to MATHEW because of the potentially strong influence of terrain on the flow configuration.

ADPIC is a Lagrangian-Eulerian transport and diffusion model. It uses the particle-in-cell method to move and diffuse marker particles in the MATHEW flow fields. ADPIC accounts for wind direction and speed shears, variable topography and surface roughness, radioactive decay, variable source rate, and effective release-stack height. A supporting code uses the output from ADPIC to calculate surface air concentrations at locations in the grid that match the locations of the actual samplers.

For the MATHEW/ADPIC calculations the wind reports used in creating the gridded three-dimensional wind fields were obtained from the TMI meteorological tower and several surface-wind reporting stations (Fig. 1). During the purge the only nearby upper-air wind measurements were taken near Middletown.

The MATHEW/ADPIC calculations were executed from several hours before the RSI sampling to 1 hour after all sampling had been completed. The early start was necessary to allow the TMI ^{85}Kr plume to reach sampling locations before sample collection began. The computer-generated surface air concentrations for the sampler locations were time-integrated every 30 minutes during the calculations. Since the RSI samplers were turned on and off at different times, both Sterling's and Bessel's central difference interpolation methods (6) were used to estimate the model concentrations for the actual sampling periods.

The RSI measured and the LLNL calculated surface air concentrations are given in Table 1 for each sampling location, listed in order from north to south. The station averages, covering sampling periods of about six hours, tend to reduce timing errors when the calculated three-dimensional transport might be slightly slower or faster than the actual transport.

The highest concentrations of ^{85}Kr were observed at and calculated for the nearest RSI sampling locations, Lititz and Brickerville, which lie east of TMI. The single highest measured concentration, 1860 pCi/m^3, was observed at Lititz during the first 2-hour sampling period. The single highest calculated concentration, 6580 pCi/m^3, was also obtained for Lititz, but not until the second 2-hour period. Concentration isopleths generated by the model (not shown) indicate that the concentration gradient at Lititz was large during most of the sampling. This accounts for the large computed concentrations during the second and third sampling periods. The model indicated higher concentrations than the measured values for the northermost sampling locations (Brickerville and Heidelburg) during the initial 2-hour period.

During the past several years, the MATHEW/ADPIC models have been evaluated against measurements from planned tracer tests (5). In September 1972 methyl iodine was released and sampled by the National Oceanic and Atmospheric Administration at the Idaho National Engineering Laboratory, Idaho Falls, Idaho. The duration of the release was three hours. In June 1974 argon-41 releases from the Savannah River Plant, Georgia were sampled by automobile for 10 minutes at each stop. In addition a sampling helicopter from EG&G, Inc., Las Vegas, Nevada, measured plume concentrations at three to seven minute intervals. The results of those studies, as well as the present comparison with the ^{85}Kr purge, are shown in Fig. 2. Half of the 2-hour RSI tracer measurements agreed with calculations within about a factor of 2.6. Possible reasons for the 2-hour curve falling to the left of the other model evaluations are the shortness of the RSI sampling interval, the lack of a reasonably dense meteorological network east of TMI (leading both to timing errors and errors in calculation of plume transport direction), and greater experimental uncertainties for some of the ^{85}Kr measurements. These experimental uncertainities have not been included in Fig. 2. The 6-hour comparison, representing averages of the three measurements and the corresponding MATHEW/ADPIC results for each location is similar to the results obtained for the prior tracer studies. Half of the calculated and experimental results agree within about a factor of two and all agree within a factor of five.

Assuming an average ^{85}Kr concentration of 200 pCi/m^3 downwind at distances of 36 to 63 km from the reactor, the skin dose rate would be about 0.04 μrem/hour (7). This rate is considerably less than the whole body dose rate of about 10 μrem/hour from the natural background of cosmic rays and radionuclides in the earth's crust. Since ^{85}Kr decays by emission of β-particles with only a 0.4% probability of γ-rays, the whole body dose from ^{85}Kr is less than one percent of the skin dose.

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Figure Legends

Fig. 1.   Locations of meteorological stations and RSI samplers during the ^{85}Kr purge of the TMI reactor. The axes are labeled in Universal Transverse Mercator coordinates. Seven sampling sites were downwind from the reactor; one was placed upwind to assess background concentrations.

Fig. 2.   Percentage of cases when ^{85}Kr concentrations calculated by MATHEW/ADPIC are within a factor N of the RSI measured concentrations, where N is the larger result, either measured or calculated, divided by the smaller result. Curves from previous tracer tests using ^{131}I and ^{41}Ar are also shown.
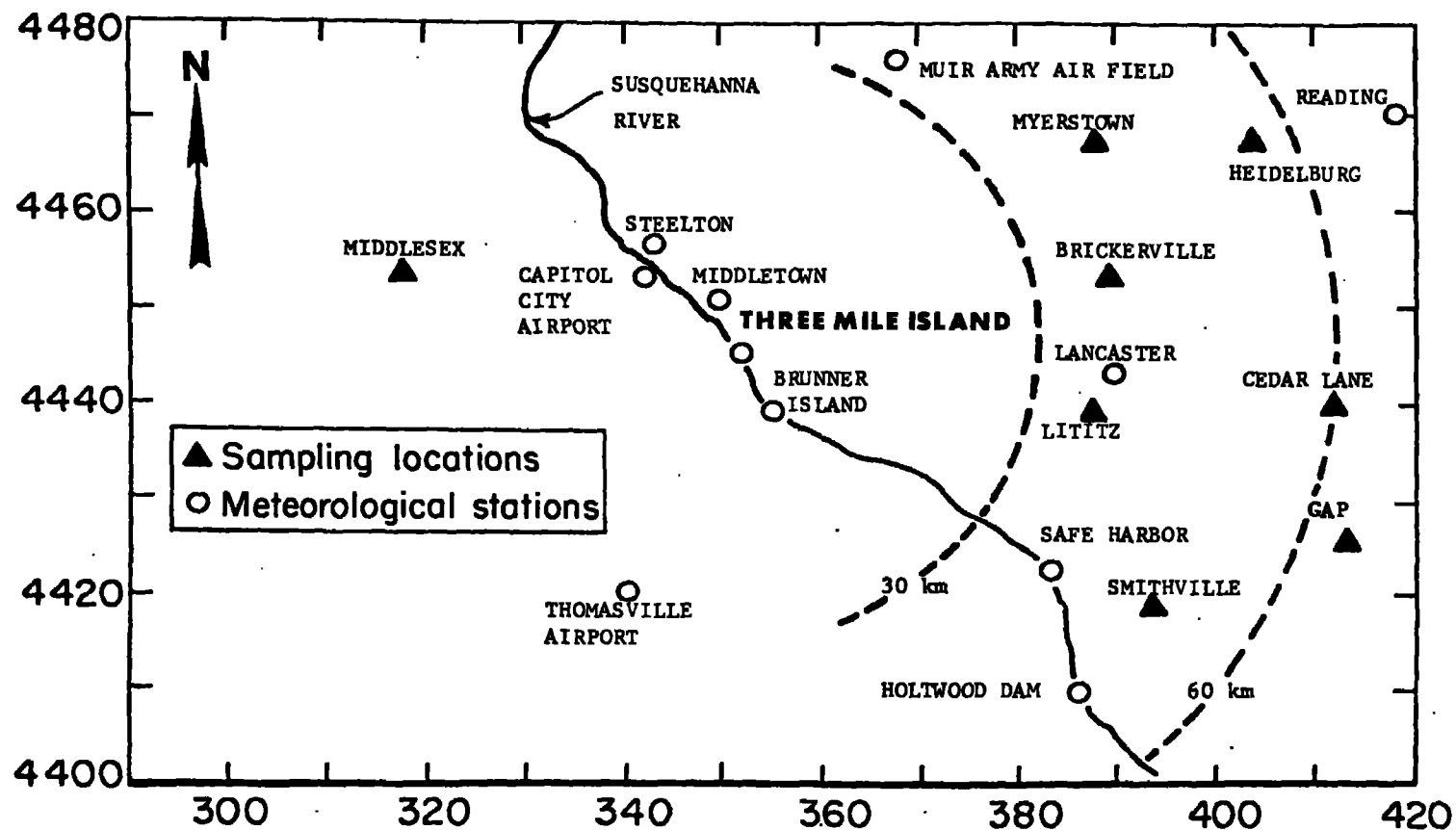
FIGURE 1. Locations of meteorological stations and RSI samplers during the ^{85}Kr purge of the TMI reactor. The axes are labeled in Universal Transverse Mercator coordinates. Seven sampling sites were downwind from the reactor; one was placed upwind to assess background concentrations.
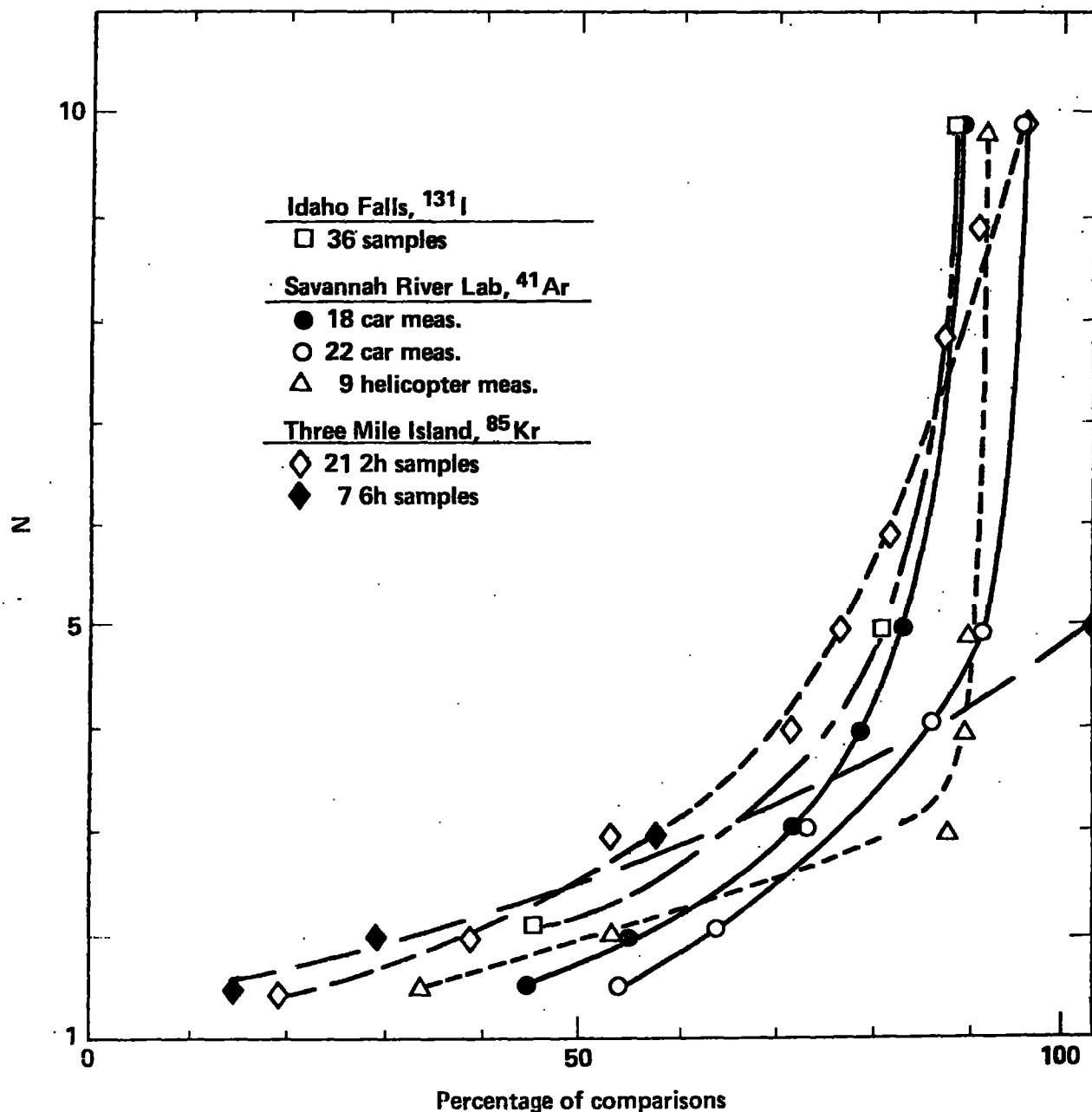
FIGURE 2. Percentage of cases when ^{85}Kr concentrations calculated by MATHEW/ADPIC are within a factor N of measured RSI concentrations, where N is the larger result, either measured or calculated, divided by the smaller result. Curves from previous tracer tests using ^{131}I and ^{41}Ar are also shown.

Table 1. RSI measured and LLNL calculated ^{85}Kr concentrations (pCi/m^3) in surface air on 8 July 1980. The values calculated by MATHEW/ADPIC include a background of 20 pCi/m^3.

Location	Distance from TMI (km)	Sampling time EDT	^{85}Kr concentration (pCi/m^3)			
			For each interval		Average	
			Measured*	Calculated	Measured*	Calculated
Meyerstown	42	1510–1715	32±30	39	48±33	26
		1720–1915	<32	20		
		1920–2100	81±36	20		
Heidelburg	55	1540–1740	<28	89	36±35	43
		1745–1945	36±32	20		
		1950–2115	<45	20		
Brickerville	37	1445–1650	830±50	1740	299±36	648
		1700–1840	36±31	185		
		1845–2040	<30	20		
Cedar Lane	60	1638–1850	<70	20	107±42	22
		1855–2130	<30	20		
		2135–0016	220±26	27		
Lititz	36	1430–1640	1860±112	648	763±61	3180
		1645–1825	200±36	6660		
		1830–2030	230±32	2220		
Gap	63	1554–1812	<67	20	<42	20
		1815–2025	<34	20		
		2030–2343	<25	20		
Smithville	48	1510–1730	<70	20	84±44	20
		1738–1948	147±26	20		
		1950–2310	<36	20		
Middlesex	34	1700–1900	<22			
		1900–2100	<22			

*Errors are ± 2 standard deviations of the counting statistics and background determinations.

References and Notes

1. E. Bretthauer, F. Grossman, W. Efurd, G. Douglas, A. Smith, and M. Bills, Environmental Radioactivity at the TMI-Venting Phase, U.S. Environmental Protection Agency unpublished manuscript, Las Vegas, NV and Washington, DC (1980).

2. The computer calculations for this study incorporated the measured source rates, the estimated momentum plume rise (from measured stack exit velocities), and verified meteorological data.

3. C. J. Paperiello, in Noble Gases (CONF-730915, ERDA, 1973) pp. 239-248.

4. C. A. Sherman, J. Appl. Meteor., 17, 312-319 (1978).

5. R. Lange, J. Appl. Meteor., 17, 320-329 (1978).

6. J. B. Scarborough, Numerical Mathematical Analysis, 5th Edition, The Johns Hopkins Press, Baltimore, MD (1962).

7. National Commission on Radiological Protection, Krypton-85 in the Atmosphere-Accumulation, Biological Significance, and Control Technology, NCRP Report No. 44, National Council on Radiation Protection and Measurements, Washington, D.C. (1975).

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